

K_a For a NaI Detector Using Two Methods

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and Frank S. Moore**

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ABSTRACT

The Analytical Development Section of SRTC was requested by the Facility Decommission Division (FDD) to determine the holdup of highly enriched uranium (HEU) in the 321-M facility as part of an overall deactivation project of the facility. The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. The facility also includes the 324-M storage building and the passageway connecting it to

321-M. The results of the holdup assays are essential for determining compliance with the Solid Waste's Waste Acceptance Criteria, Material Control & Accountability, and to meet criticality safety controls. Two measurement systems will be used to determine HEU holdup: One is a portable EG&G Dart system that contains Gamma-Vision software to support a Multichannel Analyzer (MCA) card, high voltage power, and space to store and manipulate multiple 4096-channel γ -ray spectra. The other is a 2"x 2" NaI crystal with an MCA that uses a portable computer with a Canberra NaI+ card installed. This card converts the PC to a full function MCA and contains the ancillary electronics, high voltage power supply and amplifier, required for data acquisition. This report will discuss the determination of the area-source calibration constant for a Canberra 2"x 2" NaI detector (serial number 06957694) using two methods. One method uses a calculation of the ^{235}U area-source efficiency calibration constant for a NaI detection system from a non-traditional area-source data acquisition configuration. The second method uses the traditional area-source data acquisition configuration. We present arguments that our non-traditional method is superior, and we suggest the appropriate experiment to test our arguments.

The adopted area-source calibration constant from our calculations is $(2.98 \pm 0.75) \times 10^{-6}$ g-min/in², which has been used in multiple area-source configuration holdup measurements in the deactivation of Building 321-M. A comparison of the derived constant with the calculated value of 1.20×10^{-6} g-min/in² obtained from the traditional determination of the area-source constant is provided.

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K_a For a NaI Detector Using Two Methods

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1. INTRODUCTION

Facility Decommission Division (FDD) has requested technical assistance from the Analytical Development Section (ADS) of the Savannah River Technology Center (SRTC) to determine the holdup of enriched uranium in the 321-M facility as part of an overall deactivation project of the facility.¹ The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. The facility also includes the 324-M storage building and the passageway connecting it to 321-M. The project includes the dismantling and removal of all held-up highly enriched uranium (HEU) to the extent practical. ADS was tasked with conducting holdup assays to quantify the amount of HEU on all components removed from the facility prior to placement in B-25 containers. The results of the holdup assays are essential for determining compliance with the Solid Waste's Waste Acceptance Criteria, Material Control & Accountability, and to ensure that criticality safety controls are not exceeded.

The facility operated for 25 years. During this time thousands of uranium-aluminum-alloy (U-Al) fuel tubes were produced. After the facility ceased operations in 1995 all of the easily accessible U-Al was removed from the building, and only residual amounts remained. The bulk of this residue is located in the equipment that generated and handled small U-Al particles and the exhaust systems for this equipment (e.g., Chip compactor, casting furnaces, log saw, lathes A & B, cyclone separator, Freon™ cart, riser crusher, ...etc).³

²³⁵U holdup measurements were performed in 1995 and documented in technical report WSRC-TR-95-0492.⁴ The holdup values reported in WSRC-TR-95-0492 were only best estimates, due to lack of time for conducting the measurements and analysis. In the FDD deactivation of the facility, ADS is conducting more detailed nondestructive analyses (NDA). ADS researchers are performing the recent NDA assays using a portable high purity germanium γ -ray detection system⁵ and using a portable NaI detection system.⁶ The former system was calibrated in the traditional point-, line-, and area-source configurations,⁵ however the latter was efficiency calibrated in only the point-source configuration and in an empirical close-contact configuration.⁶

Subsequent to the NaI efficiency calibrations, holdup measurements of ^{235}U in multiple 321-M process components have required use of the area-source acquisition configuration.⁽⁷⁻⁹⁾ From the NaI data available, we have calculated and estimated the required area-source calibration factors and effective fields of view as required in the NDA holdup measurements. This report describes and documents those determinations.

2. EXPERIMENTAL

We perform four distinct determinations of an area-source calibration factor for the NaI system with a 1-inch recess in the steel shield. Each uses a set of assumptions to interpret the data. The first calculation involves data that were acquired in the close-contact configuration in reference 6. These data were used in reference 6 to develop an empirical Deming least squares fit to the close-contact data. The data were clearly acquired in what would qualify as an area-source configuration in which the detector is viewing a constant unit area of the source. We use our point-source configuration to determine what portion of the source the detector is able to view in the case of each acquisition. The detector crystal is a right circular cylinder of radius 1 inch. The shield is a right circular cylinder of radius $1\frac{3}{16}$ inches. We therefore assume the detector is able to view exactly 1.41π square inches and determine a self-absorption corrected calibration factor K_a for the detection system. The close-contact acquisition configuration is sketched in Figure 1, where for an arbitrary source the ^{235}U distribution extends well outside of the 1.41π square inches field of view of the collimated detector.

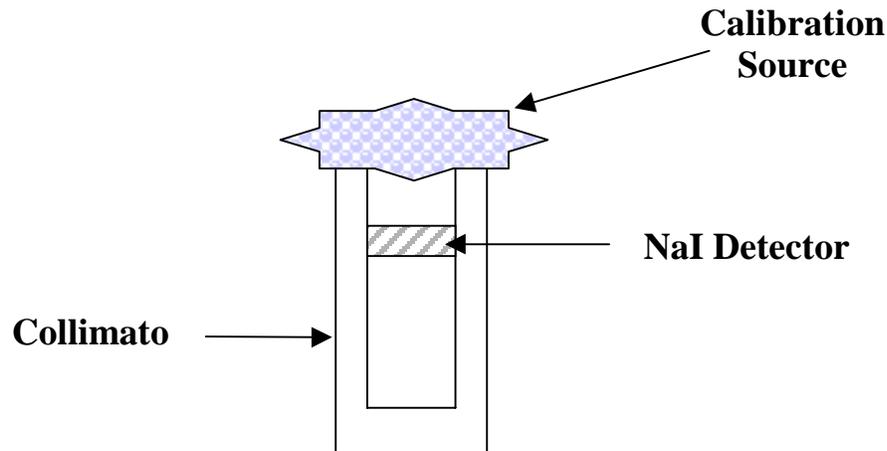


Figure 1. Sketch of the close-contact acquisitions.

2.1 Non Traditional Method

2.1.1 Determination of K_a from Close-contact Data

It seems the best explanation of our process to calculate an individual K_a from the contact measurements is to proceed by an example calculation. From reference 6 we determined a point source calibration factor of $K_p = (2.10 \pm 0.36) \times 10^{-6}$ g-min/in² for our NaI detection system. This calibration factor was determined from two sets of acquired data from ranges of 8.25 inches and 8.5 inches. We then acquired close-contact data from each of fourteen sources with pre-determined self-absorption correction factors.^{5,6} The close-contact data are listed in Table 1 along with the mass and transmission correction factors for each source.

In column five of Table 1 we have listed the close-contact value transmission-corrected (T-corrected) cpm/g, which for each source is simply the observed close-contact count rate multiplied by the transmission correction factor of column four and divided by the mass of the source. For sources wt2014a, wt2017a, wt2020a, and wt2024a we assume the detector is able to view the entire source in the close-contact configuration. We assume that the detector is viewing only a portion of all of the remaining ten sources. For the four full-view sources listed, we obtain an average T-corrected cpm/g value of 99000 ± 23000 . We then use this value to estimate what portion of each of the other sources the detector is able to view in the close-contact configuration.

In our example calculation, we use source wt2015a, which has a mass of 9.90 g. The observed ²³⁵U mass is

$$\frac{\text{cpm} * \text{Cf}}{\text{cpm/g}} = \frac{480980 * 1.07}{99000} = 5.2 \text{ g}, \quad (1)$$

where the denominator is the average cpm/g value for the four full view sources.

With this technique we determine the field of view listed in column six for each source. This field of view is then representative of an infinite area-source, where the remaining portion of the source is outside of the view defined by the steel-clad shield. We use these area-source data to determine the area-source calibration constant from equation (2).

$$^{235}\text{U} = (\text{cpm})(K_a)(\text{Cf})(\text{Effective Area}). \quad (2)$$

Re-arranging

$$K_a = ^{235}\text{U}_{\text{observable}} / (\text{cpm})(\text{Cf})(\text{Effective Area}). \quad (3)$$

The Effective Area in each of the close-contact measurements is 1.41π square inches. For our source wt2015a example

$$K_a = ^{235}\text{U}_{\text{observable}} / (480980)(1.07)(1.41\pi) = 2.28(47) \times 10^{-6} \text{ g-min/in}^2.$$

For the far-field detector calibration of the NaI detector 06957694 we used a distance of 8.25" from source to detector face. We acquired spectra from fourteen ^{235}U standards ranging in mass from 0.65 g up to 98.53 g (reference 3). The spectra were acquired using the Canberra NaI+ multichannel analyzer with Genie 2000 software for control of the acquisition parameters and storage of spectra. The spectra were acquired in the energy range 0 – 2 MeV with an ADC gain of 1024 channels and with the detector high voltage set to +800 V.

A total of twenty-five spectra from the fourteen standards were acquired. Eleven were acquired in the far-field configuration from 8.25", and fourteen were acquired from the close-field configuration at 1" from the surface of the detector. These distances do not include source thickness, which for some ranged up to about 1.5". A typical spectrum is shown in Figure 2, where the 185.7 KeV peak is labeled. In each of the far-field spectra, we assumed the detector including shielding could obtain a 100% field of view of the standard. Only sample self absorption limited the interaction between the 185 KeV γ -ray from ^{235}U and the detector. In the close-field configuration the detector shielding prevented a full view of some of the larger standards. In the close-field configuration we had to consider limitations from both sample self absorption and field of view considerations. Several weeks later we repeated the far-field configuration measurements using a source to detector distance of 8.5".

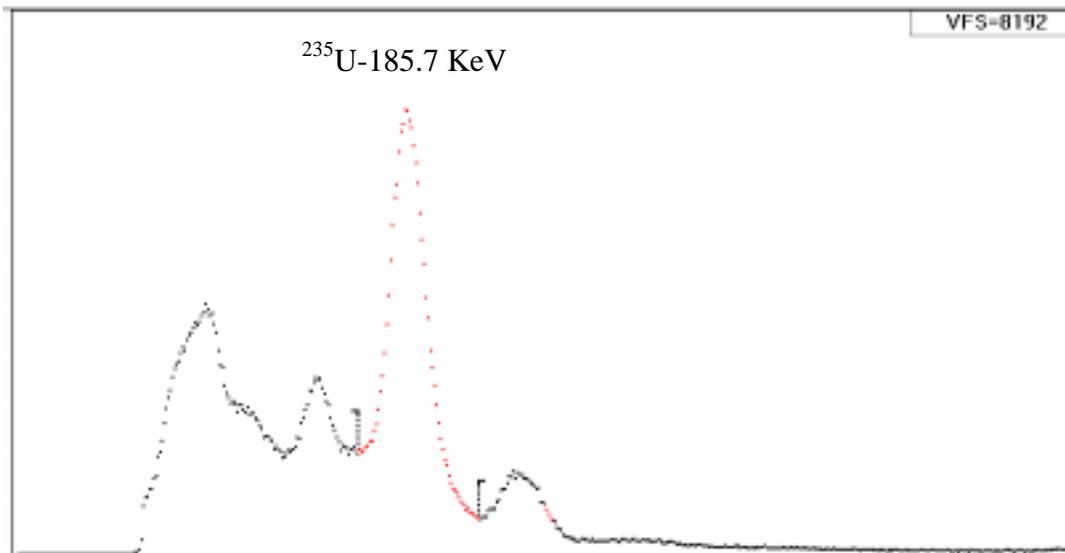


Figure 2. A typical NaI spectrum showing an ROI of the 185 KeV peak of U-235.

Table 1. A list of data from the Close-contact acquisitions. All count times are one minute.

Standard	Mass (g)	185 KeV Area (cpm)	T Correction	T _{corr'd} (cpm/g)	Viewed Mass	Calculated K _a (x10 ⁻⁶)
wt2014a	1.01	104770	1	103733	1.06±0.25	2.28±0.54
wt2015a	9.9	480980	1.07	52000	5.22±1.06	2.29±0.53
wt2016a	39.11	632996	1.5	24250	9.62±4.70	2.29±1.12
wt2017a	0.99	133740	1	135091	1.35±0.32	2.28±0.48
wt2018a	5.96	243347	1	40830	2.47±0.60	2.29±0.55
wt2019a	15.78	400147	1.15	29200	4.67±1.12	2.29±0.40
wt2020a	0.71	69212	1	97482	0.70±0.17	2.28±0.55
wt2021a	4.2	231535	1	58000	2.47±0.60	2.41±0.58
wt2022a	9.91	508545	1.07	54900	5.52±1.33	2.29±0.56
wt2023a	0.65	48729	1	74968	0.49±0.12	2.27±0.55
wt2024a	2.74	224221	1	81832	2.27±0.54	2.28±0.55
wt2025a	4.41	305430	1	69259	3.10±0.74	2.29±0.55
wt2026a	98.31	681817	2.95	20500	20.44±10.64	2.29±1.26
fl0001	98.53	578004	2.95	17300	17.28(519)	2.29±0.80

The calculated K_a for each source is listed in column seven of Table 1. The error in each is dominated by the uncertainty in the average count rate of the five full-view samples. The uncertainty in the whole technique is dominated by the error in the approximation made that each of these five full-view sources represents an area-source configuration even in this close-contact configuration. The results listed in column seven of Table 1 must be regarded as a check on our other techniques of calculation below rather than as an independent calculation of K_a. The fourteen measured values in Table 1 yield an average value for this calculation of 2.30(18)x10⁻⁶ g-min/in².

2.1.2 Determination of K_a From Extrapolated 2-, 3-, and 4-inch Recess Data

Our second determination of K_a uses data acquired at larger recesses of the detector inside the steel shield.¹⁰ We take the measured effective areas from these efficiency calibrations and extrapolate to a 1" recess to determine the effective area for the 1" recess configuration. We then determine K_a from data acquired from a distance of 8" using the area-source configuration formula of equation (2).

In reference 10 we determined the effective area for the area-source configurations in the efficiency calibration of an identical 2' x 2' NaI detector. These effective areas were determined from an acquisition distance of 24" with the detector recessed successively 2", 3", and 4" in the cylindrical shield and were determined for the Plutonium γ-rays in the energy window 283 KeV – 514 KeV. While the detection efficiency is clearly dependent upon the photon energy, we assume in this calculation that the effective area is

approximately independent of the photon energy. Thus we assume that the measured effective area for the Plutonium photons is approximately representative of the effective area for the ^{235}U photon at 185 KeV.

From reference 10 the effective areas at 24" of the 2 inch recessed detector, 3 inch recessed detector, and 4 inch recessed detector are 2157.30 cm², 1337.67 cm², and 770.57 cm² respectively. These three data points are shown in Figure 3 and define the quadratic curve

$$A(\text{recess}) = 4554.1 - 1450.9x + 126.26x^2, \quad (4)$$

where x is the recess distance in inches. For a one-inch recess and a source to detector distance of 24 inches we extrapolate an effective area of 3229.5 cm². We adjust to a source to detector distance of 8.25 inches, whence we wish to determine the K_a calibration constant by assuming the effective area varies approximately with the square of the source to detector distance. Hence

$$\begin{aligned} A(d) &= A_0(d/d_0)^2 = 3229.5(d/24)^2 = 3229.5(8.25/24)^2 & (5) \\ &= 382 \text{ cm}^2 & = 59 \text{ in}^2. \end{aligned}$$

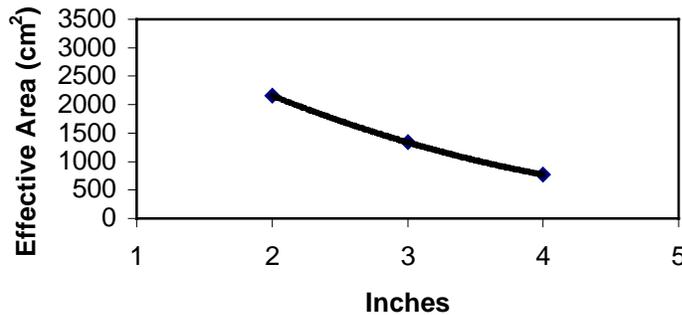


Figure 3. A Plot of Effective Area Versus Detector Recess.

We then return to the ^{235}U calibration data that were acquired at a distance of 8.25 inches and determine the area-source constant using equation (3). In references 6 and 11 we obtained a corrected detection rate of 5100(1000) cpm/g in this configuration. We calculate¹²

$$\begin{aligned} K_a &= \frac{^{235}\text{U}}{(\text{cpm})(Cf)(\text{effective area})} \\ (3) &= \frac{1}{(5100)(1)(59)} = 3.31(65) \times 10^{-6} \text{ g-min/in}^2, \end{aligned}$$

which is in fairly good agreement with our approximation above. A second calculation from data acquired at 8.5 inches with an effective area of 63 in² yields a value of 2.40(41)x10⁻⁶ g-min/in².

2.1.3 Theoretical Calculation of ²³⁵U K_a from Experimental Pu Values

Our fourth approximation comes from a theoretical calculation of the ²³⁵U calibration constant from the experimental ²³⁹Pu area-source constant. To do this we adjust the detection rates for the ²³⁹Pu γ-rays in the detection window of 283 KeV – 514 KeV to the detection rates for the ²³⁵U γ-ray in the 185 KeV detection window. We must account for the differences in half-life, branching ratios, and energy dependence of the detection rate.

The ²³⁹Pu area-source constants were determined experimentally for an identical 2” x 2” NaI detector at detector recesses of 2, 3, and 4 inches to be 2.19x10⁻⁵ g-sec/cm², 3.65x10⁻⁵ g-sec/cm², and 6.27x10⁻⁵ g-sec/cm² respectively. These three data points are shown in Figure 4 and define the quadratic curve

$$K_a(\text{recess}) = [2.75 - 1.44x + 0.58x^2]10^{-5}, \quad (6)$$

where x is the recess distance in inches. Extrapolating to 1 inch yields a Pu calibration constant of 1.89x10⁻⁵ g-sec/cm² or 2.05x10⁻⁶ g-min/in², which we then wish to transform to a ²³⁵U calibration constant.

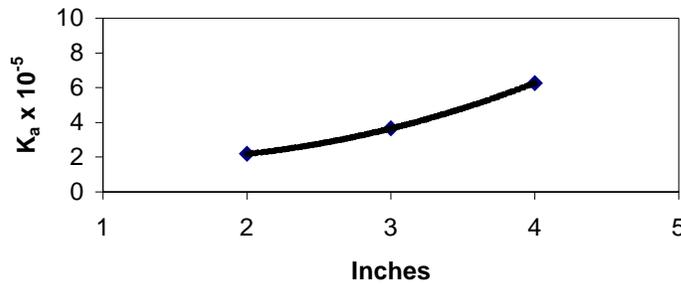


Figure 4. K_a for ²³⁹Pu Versus Detector Recess.

We first adjust for the difference in decay half-lives. This ratio is (7.04x10⁸/24110) = 29200. We then correct for the difference in branching ratios of the photons that each calibration constant represents. The branching ratio for the ²³⁵U decay at 185 KeV is 0.53. The branch for the ²³⁹Pu photons is the sum of the branching ratios for all of the γ-rays in the window 283 KeV – 514 KeV. From reference 13 that sum is 0.00006251.

Transforming, we obtain

$$K_a' = K_a(29200)(0.00006251/0.53) = 7.06x10^{-6} \text{ g-min/in}^2, \quad (7)$$

which we still must adjust for the energy dependence of the detection efficiency. From Figure 12-4 of reference 14 we take that energy dependence to be 1.8. Finally we obtain the estimate

$$K_a^{235\text{U}} = K_a'/1.8 = 3.92 \times 10^{-6} \text{ g-min/in}^2. \quad (8)$$

As a check on this calculation we can apply the same reasoning and corrections to transform the ^{239}Pu point source calibration constant determined in reference 10 to a ^{235}U point source calibration constant. This transformation requires no knowledge or measure of the detector field of view. The point source constant for ^{239}Pu determined in reference 10 is $1.10 \times 10^{-5} \text{ g-sec/cm}^2$. Transforming with the same factors we obtain

$$\begin{aligned} K_p^{235\text{U}} &= (1.10 \times 10^{-5})(29200)(0.00006251/0.53)(1/1.8) \\ &= 2.10 \times 10^{-5} \text{ g-sec/cm}^2 = 2.26 \times 10^{-6} \text{ g-min/in}^2, \quad (9) \end{aligned}$$

in extremely good agreement with the calculated K_p of $(2.10 \pm 0.36) \times 10^{-6}$.

2.2 Traditional Method of Determination of K_a And Effective Area

Subsequent to performing the assays of references 7 – 9 and subsequent to performing the calculations of this report, we performed the traditional line and area-source efficiency calibrations of the NaI detection system using a 1" recess of the detector in the steel shield. The measurements were obtained using the 4.41 g ^{235}U source number wt2025a at a distance of 40 inches and using a lateral increment of six inches. The measurement is diagramed in Figure 5 and uses the method prescribed in reference 15.

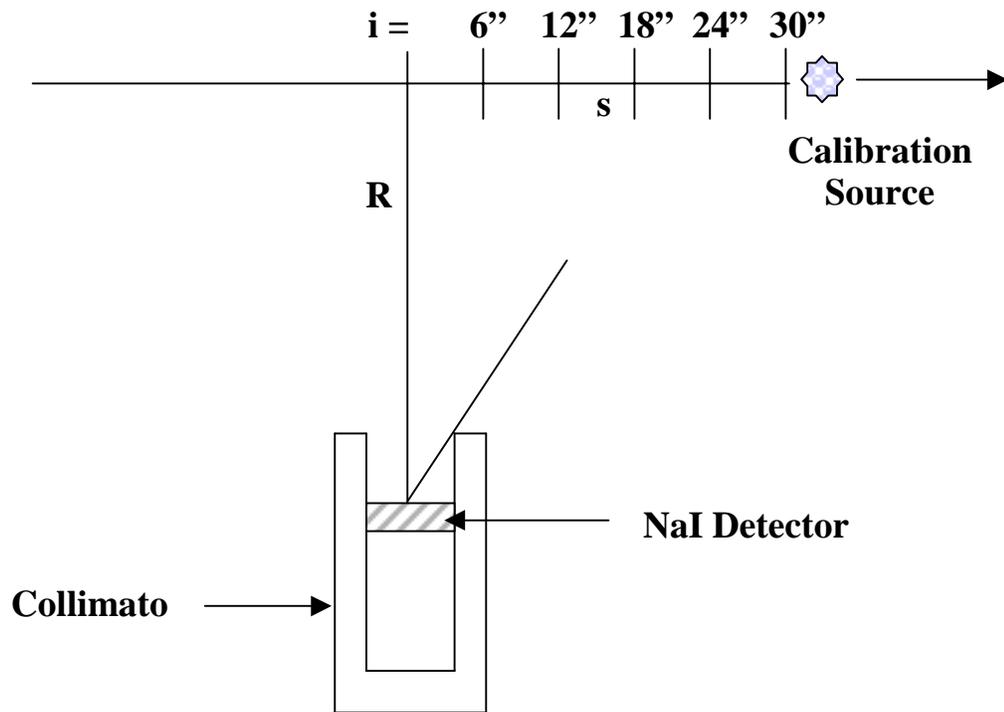


Figure 5. Sketch of the Geometric Calibration Setup.

The data of the traditional measurement are listed in Table 2. The first measurement in row one is the measurement taken with the detector viewing the 4.41 g standard straight on from a distance of 40 inches. This measurement was acquired at what we call location 0. All subsequent acquisitions were obtained by moving the standard a distance of six inches along the line perpendicular to the detector axis, as shown in Figure 5. We denote the location in the first column of Table 2. The second column lists the count times, which were all 60 seconds. The third column is the peak area, the fourth column is the uncertainty in percent of the peak area, and the fifth column is the measured counts per second in the peak. Column eleven is the calculated value of location number times measured cps of column five.

The measured K_p , effective length L , K_l , effective area A , and K_a are calculated according to the techniques described in references 5, 6, and 10. The calculated values by the traditional method for K_p are 1.91×10^{-5} g-sec/cm² or 2.05×10^{-6} g-min/in², which is in very good agreement with the value calculated in reference 6. The traditional method value for K_a is 1.118×10^{-5} g-sec/cm² or 1.20×10^{-6} g-min/in². Using the traditional method we calculated an effective area at 40" and 1" recess of 17636 cm². These values contrast with the values predicted by our four techniques by nearly the same ratio, as we discuss immediately below.

The average of our four calculations of K_a is $2.98(78) \times 10^{-6}$ g-min/in². The ratio of the calculations to the traditional measurement of K_a is $(2.98 \pm 0.78) / 1.20 = (2.48 \pm 0.65)$, where we assume all of the uncertainty is in the numerator. The ratio of measured effective area to extrapolated effective area at 40" and 1" recess is $17636 \text{ cm}^2 / 8971 \text{ cm}^2 = 1.97$, where the numerator comes from Table 2 and the denominator comes from equation (5) with $d = 40$, and $d_0 = 24$. Therefore the entire difference between our calculated area-source calibration constant and the traditional measured area-source constant is accounted for by the difference between the calculated and measured effective areas.

Table 2. A list of the Traditional Data and K_p , K_l , and K_a Calculations.

Location	Count Time (sec)	185 KeV Area	% σ 185 KeV Area	cps	% σ	K_p (g-sec/cm ²)	L (cm)	K_l (g-sec/cm ²)	K_a (g-sec/cm ²)	i* C_i
0	60	1342	5.4	22.36667	0.00402385	1.91E-05	126.9849	1.52824E-05	1.11802E-05	
1	60	1337	5.4	22.28333	0.00403889					22.28333
2	60	1091	5.77	18.18333	0.00528873					36.36667
3	60	771	7.8	12.85	0.01011673					38.55
4	60	491	11.6	8.183333	0.02362525					32.73333
5	60	437	12.13	7.283333	0.02775744					36.41667
6	60	413	12.03	6.883333	0.02912833					41.3
7	60	187	25.21	3.116667	0.13481283					21.81667
8	60	193	23.8	3.216667	0.12331606					25.73333
9	60	82	47.8	1.366667						12.3

Effective Area = 17635.5 cm²

= 2734 in².

3. DISCUSSION

The four calculated results we obtained for the area-source calibration constant yield an average value of $2.98(78) \times 10^{-6}$ g-min/in². The values have acceptably good precision. Considering that the calculations represent three independent approaches, we believe that the net accuracy is very likely satisfactory for NDA holdup purposes. We used the calibration constant and estimated effective areas extensively in determinations of HEU holdup in 321-M process equipment.⁷⁻⁹

Each of our four calculations suffers from a rationalized approximation. The first technique basically assumes that the five calibration sources wt2014a, wt2017a, wt2020a, wt2020a, and wt2024a that we viewed in the close coupled configuration were representative of an area-source configuration. We made this approximation even when we believe they did not fill the field of view of the detector in the close-coupled configuration and therefore could not be uniform area-sources.

The second and third calculations assume that the effective areas of the ²³⁹Pu photons are the same as the ²³⁵U photons for the collimated 2" x 2" detector. That is very likely a good assumption, considering that the two shields, though not identical, are very good at collimating these low energy γ -rays. We also assume the effective areas can be extrapolated to a 1-inch detector recess and can then be adjusted by the technique of equation (5).

The theoretical calculation used in the fourth technique is an exact calculation within the limits of the known relative decay half lives and γ -ray branching ratios of ²³⁹Pu and ²³⁵U and within the limits of the relative detection efficiencies for the ²³⁹Pu and ²³⁵U γ -rays. We again assume that the smoothly varying and continuous ²³⁹Pu area-source constants at 2-, 3-, and 4-inch recesses can be extrapolated to a 1-inch recess. The other factors in the calculation come from accurately known constants and accurately measured values. Using this technique we are able to very accurately calculate the point source calibration constant K_p for U-235 from the measured point source calibration constant for Pu-239.

It is important to realize that none of the calculations, including the traditional method described in reference 15, is an exact calculation of the area-source calibration constant. The traditional technique uses a point source to model an area-source. That technique provides an excellent method to determine the effective viewing area of a detection system. However its final calculation of area-source constant simply uses the point source detection rate. That is, in reference 15

$$K_a = \text{mass}/(\text{effective area})(C_0), \quad (10)$$

where C_0 is the on-axis point source acquisition rate. That is, the first acquisition rate obtained in the experiment sketched in Figure 5.

This is not equivalent to the detection rate of the same collimated detector observing a true area-source. The off-axis measurements in the traditional calculation are made only to obtain a measure of the effective viewing area of the detection system. The author of reference 15 makes the point that the effective viewing area measurements can be made with a completely different source. This statement serves to emphasize the point that ultimately the approximation of K_a comes from a single point source observation.

A true measure of area-source calibration constant can come only from the observed rate C_{planar} in equation (10) obtained from the detection system viewing a planar source of known mass/unit area. That is

$$K_a = \text{mass}/(\text{effective area})(C_{\text{planar}}), \quad (11)$$

For comparison, we take the example of our 4.41-g mass of U-235 used in the traditional measurement of section 2.2. After determination of the effective viewing area, if we calculated K_a from C_{planar} instead of C_0 , the value of K_a would increase toward our calculated values of section 2.1. That is, if the decay source were spread out over the entire effective viewing area of the detector, the detection rate C_{planar} would be lower than the detection rate C_0 , thus increasing the calculated value K_a .

For this reason the calculations of section 2.1.1 seem to us to provide the best approximation of K_a . That is, in those close-coupled acquisitions (Figure 1) the detection system would seem to be making the best uniform area-source observations. The uncertainty in those measurements is dominated by our estimation of the source mass and transmission correction factor.

4. CONCLUSION

We have used four different methods for calculation of the area-source efficiency calibration constant (K_a) for determination of ^{235}U from the detection rate of the 185.7-KeV γ -ray using our 2" x 2" NaI portable detector. The results of the four separate calculations yielded better than 27% precision. All of the four calculations yield values that are at least a factor of two larger than the value calculated in the traditional method.

We used this determination of K_a for holdup measurements in four pieces of process equipment in the deactivation and decontamination of Building 321-M. Since holdup determinations in process equipment intrinsically contain very large uncertainty (i.e. +100%, -50%), the derived 27% precision is adequate for the intended purposes.

The accuracy of the four calculations was checked by the traditional experimental technique for determination of the effective field of view and area-source calibration constant. The methods do not yield satisfactory agreement, and we present arguments that point toward acceptance of the non-traditional calculations. We propose to perform experiments using a true planar source to determine K_a using equation (11) of this report.

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